

CONSIDERING CLAY ROCK HETEROGENEITY IN RADIONUCLIDE RETENTION

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The Callovo-Oxfordien clay rock formation has a strong retention capacity for radionuclides, a favorable condition for the implementation of a nuclear waste repository. Principal retaining minerals are illite, and interstratified illite/smectite (I/S). Radionuclide retention has been studied on illite, illite/smectite and on clay rock obtained from different locations and data for retention on bentonite (80% smectite) are available. Sorption depends on the type of mineral, composition of mineralogical assemblages, individual mineral ion exchange capacities, ion distribution on exchange sites, specific surface areas, surface site types and densities for surface complexation as well as on water/rock ratios, temperature etc. As a consequence of mineralogical and textural variations, radionuclide retention properties are expected to vary with depth in the Callovo-Oxfordian formation. Using a simple additivity approach for the case of sorption of Cs and Ni it is shown that models and databases for illite and bentonite can be used to describe sorption in heterogeneous clay rock systems.

A surface complexation/ion-exchange model as proposed by Bradbury and Baeyens [1] without electrostatic contributions, was used directly as far as acid base properties are concerned but was modified [2] with respect to sorption constants, in order to describe Na-, Ca, and Cs montmorillonite and bentonite MX-80 with a single set of surface complexation constants and also to account for carbonate and sulphate concentrations in groundwater. The model is integrated into the geochemical code PHREEQC [3] considering dissolution/precipitation/solubility constraints of accessory minerals (calcite, illite, celestite, quartz). Site densities for surface complexation and ion exchange are derived from the mass fractions of illite and of smectite in illite/smectite obtained from an overall fit of measured CEC data from all samples of the EST205 drill core considering the measured mass fraction of illite, I/S, and kaolinite. Five values were fitted simultaneously in the overall fit: the CECs of the kaolinite, the smectite and illite components as well as the illite fraction in both R0 and R1 I/S.

For transport calculations of radionuclides it is important to know which part of the overall porosity belongs to the micropores and which to the interlayers of the smectite layers in the interstratified I/S. Making the hypothesis that, at the given degree of compaction, these interlayers are hydrated by two water layers one could calculate the fraction of micropores in total porosity from the measured I/S content and the fitted I/S ratio given for either R0 or R1. For high smectite contents there is about twice as much water in interlayers than in the micropores, while there is much more water in the micropores in case of low smectite contents.

The retention and porosity models for Bentonite have been adapted successfully to argillite samples for various drill cores assuming additivity of the various mineralogical contributions, in particular illite and I/S and of the illite to smectite ratio in I/S. The approach leads to a reasonable quantitative representation of the CEC as well as of surface complexation site densities as a function of depth and mineralogical composition. Ratios of micropores to interlayer pore space were obtained as well. The good correlation of calculated and measured retention data for Cs and for Ni gave reasonable justification to apply the model for the prediction of Kd variations of Cs all along a drill core. Predicted values vary by about a factor of 10.

References:

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